New 193Ir(n,n'y)193mIr Evaluated Nuclear Cross Sections for Radchem

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New ^{193}Ir(n,n'γ)^{193m}Ir Evaluated Nuclear Cross Sections for Radchem

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New measurements performed with the GEANIE γ-ray detector array at LANSCE, and theoretical calculations performed by T-16 have improved the accuracy with which the energy-dependent cross section for production of the long-lived isomer in ^{193}Ir is known. Comparisons with critical assemblies data show excellent agreement. Evaluation work is nearly complete to enable the use of the new data in applied calculations.

Introduction

The ^{193}Ir(n,n'γ)^{193m}Ir inelastic reaction provides a unique fission-neutron flux detector. The meta-stable state (or isomer) in ^{193}Ir has a 10.5-day half-life and an excitation energy of 80 keV. While many (n,2n) reactions have been used as radchem detectors to measure 14 MeV neutron fluences, and many (n,γ) reactions have been used to study low energy neutron fluences, ^{193}Ir is the only radchem diagnostic used that provides information in the few MeV region. Measuring the production of the isomer was a very difficult problem solved by Jim Gilmore, Don Barr, Moses Attrep and others at LANL.

To properly interpret neutron fluence data from radiochemical measurements using ^{193}Ir, an accurate knowledge of the isomer production cross section as a function of energy is needed. Recent measurements with GEANIE[1] at the Weapons Neutron Research (WNR) facility[2] of the Los Alamos Neutron Science Center (LANSCE), combined with theoretical modeling by T-16, have greatly improved the accuracy of this important cross section. Results are compared with data from critical assembly tests.

Experiment

Only two differential cross section measurements of ^{193}Ir isomer production have been performed. The measurements of Bayhurst *et al.* in 1975 at four incident neutron energies above $E_n = 7.5$ MeV using the activation technique[3], and the recent GEANIE measurements of the γ rays feeding the isomeric state. The new indirect measurement with GEANIE covers the entire energy range of interest and with greater precision than the only previous measurements, with the only exception being the 14 MeV data point of Bayhurst, *et al.*

GEANIE, at the LANSCE/WNR facility, is an array of high-resolution germanium detectors. Use of the time-of-flight technique allows determination of the incident neutron energy for the broad spectrum of neutrons produced by the WNR spallation neutron source. The high-energy resolution for the γ rays allows identification and yield determination for individual γ rays.

We observed 64 γ rays in ^{193}Ir, 8 of which had not been observed before. Twenty-three of these γ rays populate the isomeric state at 80 keV, either directly or by cascade. Four of the 23 γ rays are strong γ rays that populate the isomer independently, i.e. not in cascade. These 4 discrete lines have energies of 219.2 keV, 389.1 keV, 398.8 keV and 483.2 keV. The yield of each of these γ rays was measured as a function of the incident neutron energy.

The absolute normalization for the cross sections for production of individual Ir γ rays was determined using a value for the ^{nat}Fe(n,n'γ) cross section for the 847-keV γ ray at $E_n = 14.5$ MeV of 705 mb. This was determined from absolute measurements using our ^{235}U/^{238}U fission chamber and agrees well with measurements relative to ^{nat}Cr(n,n'γ) 1434-keV γ ray that we have performed using the evaluated cross section at 14.5 MeV of 695 (+/-35) mb from Simakov, *et al.*[4] We consider the Cr(n,n'γ) 1434-keV cross section to be a reliable standard near $E_n = 14$ MeV.

In obtaining the absolute cross sections, corrections were made for γ–ray attenuation in the sample. This correction was a factor of 1.85 for the low-energy 219.2-keV γ ray in the LEPS detectors. Detailed Monte Carlo modeling of the array with MCNP[5] was used for these calculations. A level diagram showing the observed γ–ray decays to the first excited state is shown in Fig. 1. Plots showing three evaluations, the previous data, and the new data, adjusted with the GNASH[6] calculations, are given in Fig. 2. A full report of the measurement will be given in a Los Alamos report that is being prepared by N. Fotiades, *et al.*[7]
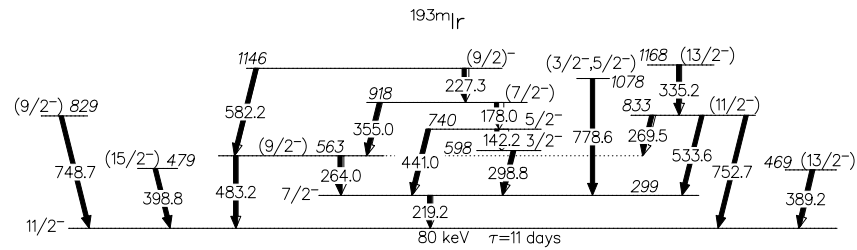
Fig. 1. Gamma rays feeding the 80-keV isomeric state in ^{193}Ir observed in the GEANIE measurement are shown with energy levels, spins and parities.

Theoretical Model Calculations

The GNASH code that incorporates statistical Hauser-Feshbach compound-nuclear modeling with pre-equilibrium and direct processes was used to calculate the cross sections for production of the observed γ rays and to correct for the unobserved, direct (from the ground state to the isomer) population of the isomer and for weaker unobserved γ rays. GNASH relies on coupled channels optical model calculations to infer elastic and reaction cross sections, and to provide transmission coefficients values to be used in the Hauser-Feshbach decay equations. The optical model potential of P. G. Young determined from fitting total cross section and other data were used. In the coupled channels calculation, the first three members of the ground state rotational band are explicitly coupled. The nuclear level densities used were of the Ignatyuk prescription that includes a "washing-out" of shell effects in the level density with increasing energy. The γ–ray strength function employed is the generalized Lorentzian strength-function formalism of Kopecky-Uhl, with E1, M1, and E2 radiations included.

In applying the GNASH calculations, first the model calculations were validated through comparison with data, and an adjustment was made to better reproduce the observed γ–ray partial cross sections. Second the model predictions were used to correct the experimental data for the non-measured contributions. Figure 3 shows the experimental and calculated sums of the four strongest γ rays feeding the isomer. The new evaluation of the cross section data takes into account both data sets providing reduced uncertainties over the entire energy range of interest.

The GNASH calculations are used to add the unobserved partial cross sections to the sum of the four observed γ ray cross sections. Details of the GNASH modeling and calculations will be given in a Los Alamos report by N. Fotiades, *et al.*[7]
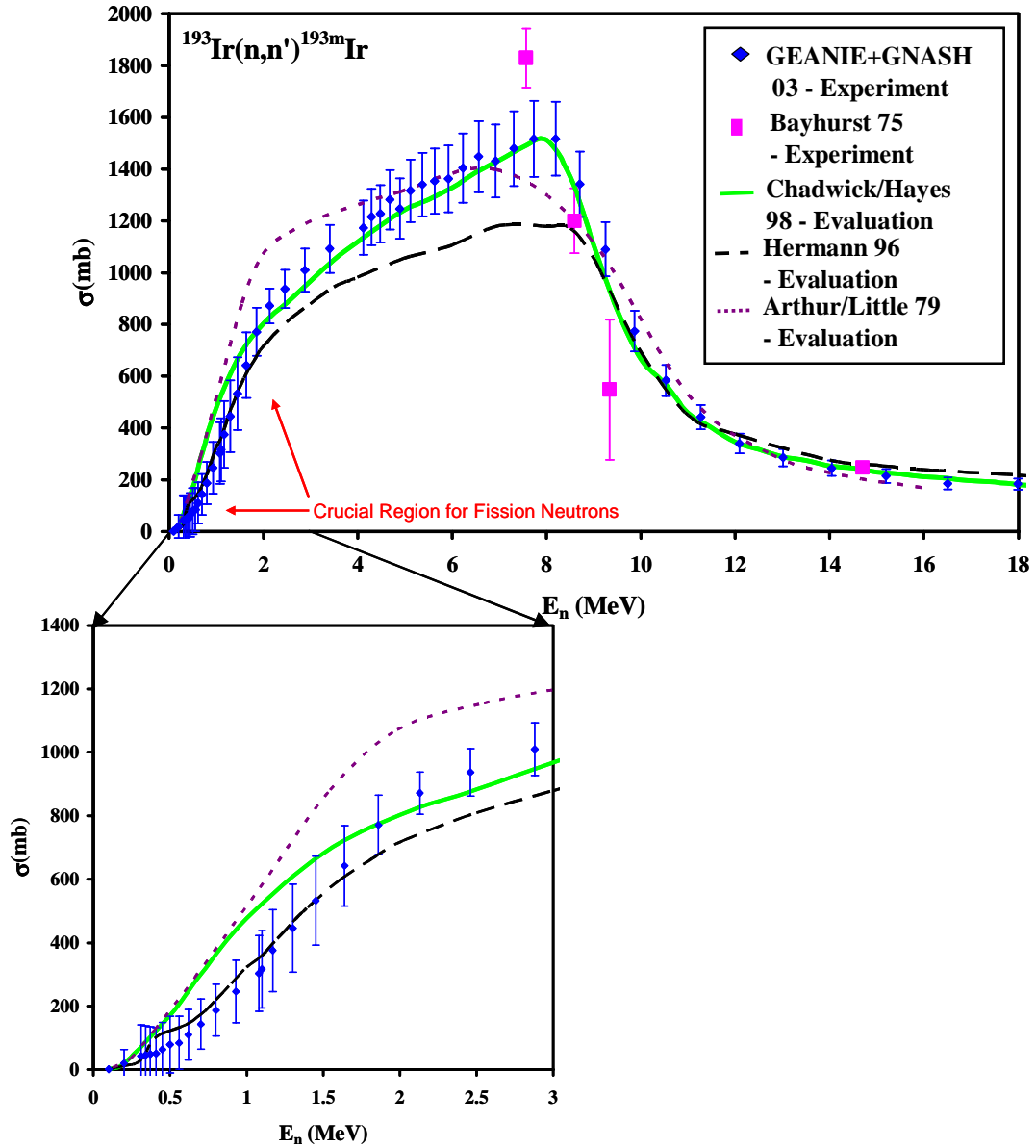
Fig. 2. The new evaluation is shown as small points with error bars, the Bayhurst data[3] as filled squares. The evaluations are: Y21GRP[8] (dotted line), Hermann[9] (dashed line) and Hayes and Chadwick[10] (solid curve). The inset shows the threshold region that is important for fission neutrons.
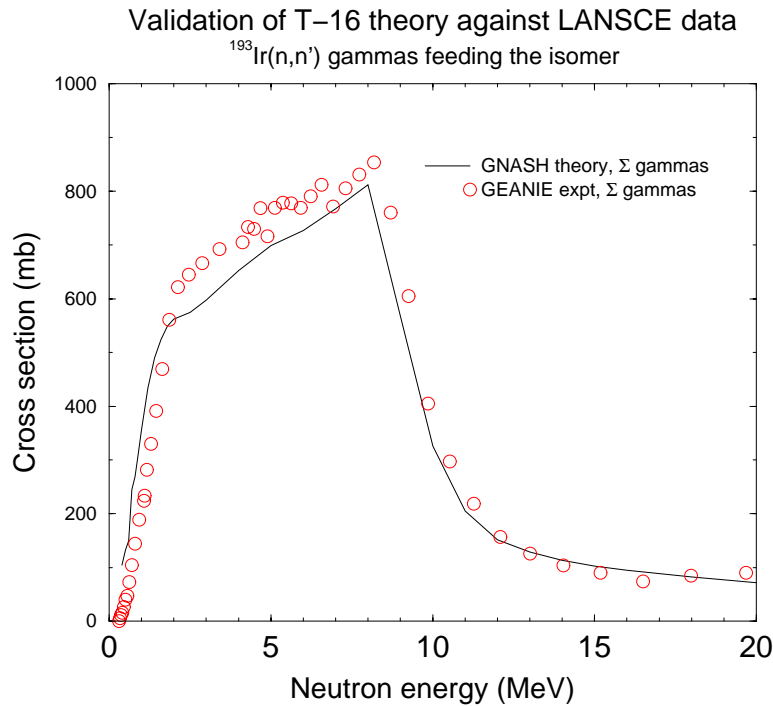
Fig. 3. The new GEANIE data (red points) for the sum of the four strong parallel γ rays are shown with the GNASH calculated sum of the same four γ rays (black line). Agreement is fairly good.

Validation with Critical Assemblies Data

Because there are undetermined constants (related to detector efficiency and branching ratios) in the Ir activation measurements, there is a normalization factor that must be determined experimentally. This normalization is determined from critical assemblies isomer production data and calculations using the cross section evaluations. Activation data are acquired for spectra of different hardness (critical assemblies provide fission neutron energy spectra that vary in hardness depending upon the makeup and configuration of a particular assembly and the location of the radchem detector within or near the assembly). The normalization factor is determined by comparing values calculated using the Ir isomer production and other necessary cross-section data, and normalizing to the critical assemblies data. These calculations usually are done using the MCNP Monte Carlo code and the SUPER[11] data library for the Ir cross-section data.

In Fig. 4, the ratio of the [193]Ir isomer production to the ^{192}Ir produced in critical assemblies tests is plotted against the ratio of the [238]U to [235]U fission yields. The fission yield ratio provides a measure of the hardness of the neutron spectrum.

We performed a simple calculation using the Y21GRP (a multi-group data set based

on SUPER) and new evaluations with critical assembly spectra to determine the normalizations and to compare with the integral data, The new evaluation gives excellent agreement using a normalization factor of 1/2.0.

The 1986 Ir cross-section evaluation (Y21GRP) of Arthur, Little and Seamon[8], when used to normalize the measured radiochemical production data to the calculated production, gave a factor of (1/2.75).

If the cross section does not have the proper energy dependence, then the shape of the calculated curve shown in Fig. 4 will not match the experimental data. This was the case for the Y21GRP evaluation, which led M. MacInnes[12] to suggest a reduction of the

Comparison of ^{193}Ir(n,n') Cross Section Evaluations Using Critical Assemblies Data

- ...•... Experimental data fit
- —□— Current X-5 evaluation divided by 2.75
- —▲— New evaulation divided by 2

Y-axis: ^{238}U/^{235}U fission yields (represents spectrum "hardness")

X-axis: ^{193m}Ir/^{192}Ir

Processed data represents Ir cross sections averaged over neutron flux spectra from traversals of critical assemblies at TA-18.

Samples of natural Ir within critical assembly contributed to both ^{191}Ir(n,γ)^{192}Ir and ^{193}Ir(n,n')^{193m} interactions.

Ratios of Ir isotopes were measured to give ^{193m}Ir/^{192}Ir ratio; unkown constants in measurements require a factor of ~2 normalization.
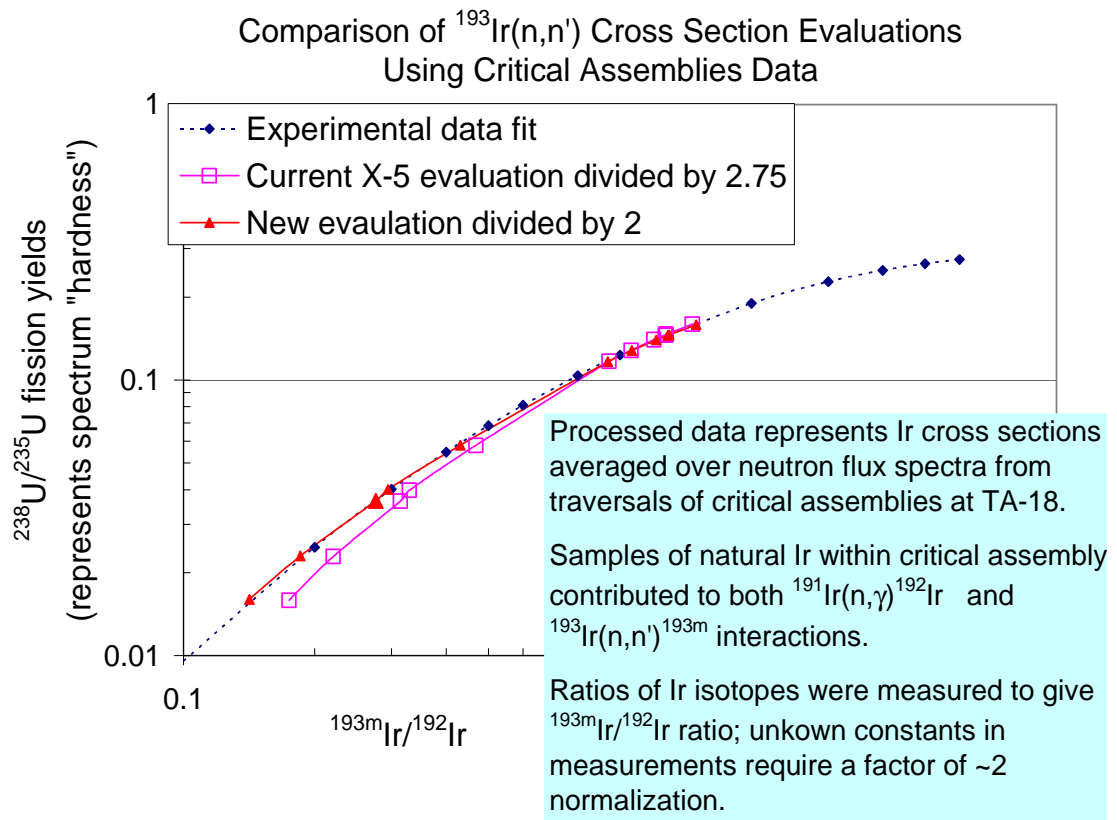
Fig. 4. Calculations using two different evaluations as input are compared with the critical assemblies experimental data.

evaluated cross section by a factor of 0.67 for incident neutron energies below 1.75 MeV. This led to a new normalization factor of (1/2.379). With this adjustment of the Ir cross-section evaluation, MacInnes achieved good agreement with the shape of the critical assemblies data. Near threshold the adjusted Y21GRP cross section data are in good agreement with the new evaluation, however at higher neutron energies there are significant differences in comparison with our new evaluation.

In the critical assemblies measurements and calculations, reactions on both the ^{193}Ir and ^{191}Ir isotopes are important. This is because natural Ir is used containing both ^{193}Ir

and ^{191}Ir. The ^{193}Ir(n,n')^{193m}Ir produces the isomer, while the ^{191}Ir(n,γ)^{192}Ir reaction produces the majority of the measured ^{192}Ir.[13]

Conclusion

The new evaluated ^{193}Ir isomer cross-section evaluation, based on recent GEANIE measurements at LANSCE/WNR and calculations in T-16 greatly improves the accuracy with which this cross section is known. Preliminary results indicate that the new evaluation is consistent with integral tests using critical assemblies. We expect that the evaluation will enhance the utility of the unique Ir neutron fluence diagnostic.

Acknowledgements

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